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Identification of New Isotopes ^{125}Pd and ^{126}Pd produced by In-flight Fission of 345 MeV/nucleon ^{238}U : First Results from the RIKEN RI Beam Factory

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A search for new isotopes using in-flight fission of a 345 MeV/nucleon ^{238}U beam has been carried out in the commissioning experiment of the next-generation in-flight radioactive isotope beam separator BigRIPS at the RI Beam Factory at the RIKEN Nishina Center. Two neutron-rich palladium isotopes ^{125}Pd and ^{126}Pd were observed for the first time, which demonstrates the great potential of the RIKEN RI beam factory.

KEYWORDS: NUCLEAR REACTION $\text{Be}(^{238}\text{U}, x) E = 345 \text{ MeV/nucleon}$, In-flight fission, New isotopes ^{125}Pd and ^{126}Pd , In-flight RI beam separator

A next-generation radioactive isotope (RI) beam facility called the RI Beam Factory (RIBF) ¹⁾ has been constructed and recently commissioned ²⁾ at the RIKEN Nishina Center that promises to advance the study of exotic nuclei to a great extent. The new superconducting in-flight RI beam separator BigRIPS is a major experimental device at the RIBF for RI beam production based on the in-flight separation technique and for research with exotic nuclei.^{3, 4)} Thanks to its large acceptances, not only projectile fragmentation of various heavy-ion beams but also in-flight fission of fissile beams ⁵⁾ can be efficiently used as a production reaction in the BigRIPS separator. Figure 1 shows a schematic layout of the BigRIPS separator along with the IRC and SRC cyclotrons¹⁾ and the ZeroDegree spectrometer.³⁾ The cyclotrons at the RIBF can accelerate all heavy ions up to approximately 400 MeV/nucleon, including very heavy elements such as uranium, with the goal of reaching an intensity of 1 pμA (6×10^{12} particles/sec).¹⁾

In-flight fission of fissile beams, such as a ^{238}U beam, is known as an excellent mechanism for producing a wide range of neutron-rich exotic nuclei far from stability. This was well demonstrated by experiments at GSI, in which more than a hundred new isotopes were identified in a single experiment.^{6, 7)} The BigRIPS separator has been designed with large acceptances, to take advantage of the high production cross sections of neutron-rich isotopes from these reactions. The full angular acceptances of 80 mrad (horizontal) and 100 mrad (vertical), and the full momentum acceptance of 6 % allows efficient RI beam production with in-flight fission, in which fission fragments are produced with large spreads in both angle and momentum. Due to its large acceptance, the BigRIPS separator has an approximately 50% efficiency for the collection of these fission fragments.

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Another important feature of the BigRIPS separator is its two-stage structure, which allows delivery of tagged RI beams, or use as a two-stage separator. In the tagging mode, which may also be called a separator-spectrometer mode, the first stage is used to produce and separate RI beams with a wedge energy degrader, while the second works as a spectrometer to analyze and identify those RI beams. The momentum resolution of the second stage has been designed to be high enough to identify RI beams without measuring their total kinetic energies, even though they are produced in several charge states in our energy domain. In the two-stage separator mode, the energy degrader is used at both of the stages in order to further purify the RI beams.

The BigRIPS separator is composed of fourteen large-aperture superconducting quadrupole triplets (STQ) and six room-temperature dipoles. They are labeled as STQ1-STQ14 and D1-D6 in Fig. 1, respectively. There are seven foci in the beam line, indicated as F1-F7. The first stage of the BigRIPS separator includes the components from the production target position (F0) to F2, while the second stage spans those from F3 to F7. The two STQs, located between F2 and F3, are used as a matching section. Each focus is contained in a focal plane chamber to accommodate various beam-line detectors used for the diagnostic and particle identification.

The commissioning of the BigRIPS separator started in March 2007. The first RI beams were successfully produced in late March using ^{86}Kr and ^{238}U primary beams. Then in a follow-up experiment in May 2007, after evaluating the performance of the ion optics in the BigRIPS separator, the detector performance and the particle identification system, some beam time was devoted to a search for new isotopes using in-flight fission of a ^{238}U beam. During these measurements we reached easily the frontiers of known isotopes and could even observe the very neutron-rich palladium ($Z=46$) isotopes ^{125}Pd and ^{126}Pd for the first time. In this paper we report on these first results from the RI Beam Factory at the RIKEN Nishina Center. Preliminary results from the experiment were briefly presented before.²⁾

The experiment was performed with a $^{238}\text{U}^{86+}$ beam that was accelerated to 345 MeV/nucleon by the cascade operation of the RIBF accelerator complex consisting of the linear accelerator, RILAC and the four cyclotrons, RRC, fRC, IRC and SRC.¹⁾ The production target was a 7 mm thick piece of beryllium. The uranium beam intensity, monitored by measuring light charged particles recoiling out of the target, was 4×10^7 particles/sec on average and $\sim 10^8$ particles/sec at most. Fission fragments emitted near zero degrees were collected and analyzed by the BigRIPS separator. No energy degraders were employed for isotopic separation in these measurements. The BigRIPS separator was used in the tagging

mode where the first stage separated the fragments by magnetic rigidity ($B\rho$) and the second stage served as a spectrograph for particle identification of the fragments. The magnetic fields in the BigRIPS separator were set according to ion-optical calculations using the code COSY INFINITY⁸⁾ that took into account the effects of measured fringe fields of the magnets. The experimental conditions to search for new isotopes, such as the target thickness and the $B\rho$ setting of the BigRIPS separator, were determined based on detailed simulations with the codes LISE++⁹⁾ and MOCADI.¹⁰⁾ The measurements were made at several $B\rho$ settings ranging from 7.0 Tm to 7.6 Tm in order to determine the production yields of neutron-rich isotopes near the known limits.

The particle identification (PID) was performed by measuring the time of flight (TOF), energy loss (ΔE) and magnetic rigidity ($B\rho$) of the fragments with the beam-line detectors. The TOF was recorded between two plastic scintillation counters placed at the F3 and F7 achromatic foci in the second stage. The scintillation counters were 0.2 mm thick and 100x100 mm² in area, and the mean flight path was 46.98 m. The ΔE was measured at F7 with two 0.35 mm thick silicon detectors with an active area of 50x50 mm². The $B\rho$ measurement was made by trajectory reconstruction from the positions and angles of the fragments measured in two position-sensitive PPACs¹¹⁾ at the F3 achromatic focus and two more at the F5 dispersive focus. The effective size of the PPACs was 240 mm (horizontal) and 150 mm (vertical). First-order ion-optical transfer matrices, which were obtained experimentally with the RI beams and the primary uranium beam, were used to reconstruct the trajectory. The $B\rho$ value of the fragments on the central trajectory (central $B\rho$) was determined by the magnetic fields of the dipoles measured by NMR probes and the central trajectory radii of the dipoles deduced from the magnetic field-map data. This method provided the high-resolution $B\rho$ measurement of each fragment as described below.

The calibration of the TOF and ΔE measurements was made using the central $B\rho$ value of the fragments. The small energy loss in the PPACs at F5 was taken into account in deducing the velocities of the fragments. The TOF values of the present measurements were around 230 nsec. The root-mean-square (r.m.s.) resolution achieved in the present TOF and $B\rho$ measurements was typically 40 psec and 0.03 %, respectively. These values are the estimates for ¹⁰⁴Zr fragments produced at a $B\rho$ setting of 7.395 Tm.

Pile-up events were rejected by comparison of the pulse-heights in the two silicon detectors at F7 and the plastic scintillation counter at F7. Inconsistent events were also excluded by constraining the beam and phase-space profiles measured by the PPACs at the foci.

Furthermore the Bp was also measured by trajectory reconstruction in the second half of the BigRIPS second stage (from the F5 dispersive focus to the F7 achromatic focus), and events with inconsistent Bp values were rejected.

The PID plots were generated by calculating the atomic number, Z, and the mass-to-charge ratio, A/Q, of each fragment from the measured TOF (velocity), ΔE and Bp values. The Z value was determined from the ΔE and TOF measurements, while the A/Q was deduced from the TOF and Bp measurements. Figure 2 shows the PID plot of Z versus A/Q that was obtained in the measurement where we searched for new isotopes. The Bp setting was 7.395 Tm and the momentum acceptance was set to $\pm 1\%$. The relative r.m.s. Z resolution was 0.55 %, and the relative r.m.s. A/Q resolution was 0.041 % for Pd isotopes.

The PID was confirmed by detecting delayed γ rays emitted from short-lived isomeric states of some of the fragments. The measurement was made at F7 by using two clover-type high-purity germanium (Ge) detectors that were placed approximately 7 cm on both sides of an 8 mm aluminum beam stopper. They were operated in an add back mode, and the overall full-energy-peak efficiency was measured to be 3.5% for 1-MeV γ rays. In the present measurement we observed delayed γ rays from ^{96}Rb that confirmed the PID. Figure 3 shows the γ ray energy spectrum gated on ^{96}Rb events in the PID plot and the time span from 0.3 to 15 μsec after the implantation. The γ ray peaks at 92.1, 116.3, 122.3, 184.8, 240.2, 300.0, 366.4, 370.0 and 461.2 keV originated from the decay of the isomeric state in ^{96}Rb with an excitation energy of 1135 keV and a half-life of 2.0 μsec .¹²⁾ The lower panel of Fig. 2 shows an enlarged view of the PID plot, in which the fully-stripped ^{96}Rb , ^{123}Pd and ^{125}Pd isotopes are labeled. The new isotope ^{125}Pd can be clearly identified. The neighboring events correspond to hydrogen-like ^{122}Pd fragments.

Figure 4 shows the projected one-dimensional A/Q spectrum of Pd isotopes, along with the two-dimensional Z versus A/Q plot expanded around the region of the new Pd isotopes. The events that did not change their charge state at the F3 and F5 PPACs were selected by gating on the reconstructed Bp spectra. The width of this gate was $\pm 1\%$ and matched the setting of the separator acceptance. The A/Q spectrum in Fig. 4 was obtained by gating the PID plot with a Z gate set to $46 \pm 0.38(1.5\sigma)$, where σ represents the absolute r.m.s. Z resolution. The peaks for the fully stripped ($Q=Z$), hydrogen-like ($Q=Z-1$) and helium-like ($Q=Z-2$) ions are well separated from each other and the new isotope ^{125}Pd is clearly identified at $A/Q = 2.717$. The peak centroid of this fully-stripped ^{125}Pd is separated by 6.2σ from the neighboring $^{122}\text{Pd}^{45+}$, where σ , the absolute r.m.s. A/Q resolution, is 0.0011. The total

yield of ^{125}Pd is 22 counts. The new isotope ^{126}Pd was also identified in both the A/Q spectrum and the Z versus A/Q plot in Fig. 4, although its total yield is only 3 counts. The integrated beam dose and the total irradiation time during the measurement were 3.6×10^{12} particles and approximately 25 hours, respectively.

Figure 5 shows the measured production rates of the fully-stripped Pd isotopes along with the predictions from the LISE++ simulation in which the production cross sections of fission fragments were calculated with the LISE++ Abrasion-Fission (AF) model.⁹⁾ This AF model relies on the so-called three excitation energy model in which three nuclei, ^{236}U , ^{226}Th and ^{220}Ra , are chosen to represent all the fissile nuclei created in the abrasion-ablation stage and then the fission fragment distribution is calculated based on the semi-empirical model of ref. 5. The standard parameters of this model, such as excitation energies and production cross sections of the fissile nuclei, that reproduced the experimental data of the $^{238}\text{U}+\text{Be}$ reaction at 750 MeV/nucleon⁷⁾ were used in the Monte Carlo mode to track the fission fragments produced in the target. The measured production rates smoothly decrease with increasing mass number, and are fairly well reproduced by the LISE++ predictions, although they are somewhat overestimated. The production cross sections of Pd isotopes have been estimated using the cross sections from the LISE++ AF model and the ratio of measured and predicted rates. The estimates for ^{124}Pd , ^{125}Pd and ^{126}Pd isotopes are 80 nb, 7 nb and 0.7 nb, respectively, and are consistent with the experimental results at GSI for ^{124}Pd (32 nb).⁷⁾

In summary, we have performed the first experiment at the RIBF using the BigRIPS in-flight RI beam separator and a search for new neutron-rich isotopes was carried out using the in-flight fission of a ^{238}U beam at 345 MeV/nucleon. We observed the production of two new isotopes ^{125}Pd and ^{126}Pd , even though the uranium beam intensity was far from the goal for the RIBF and the total observation time was only about a day. This discovery demonstrates not only the performance of the BigRIPS separator, but also marks the launch of this new-generation RI beam facility.

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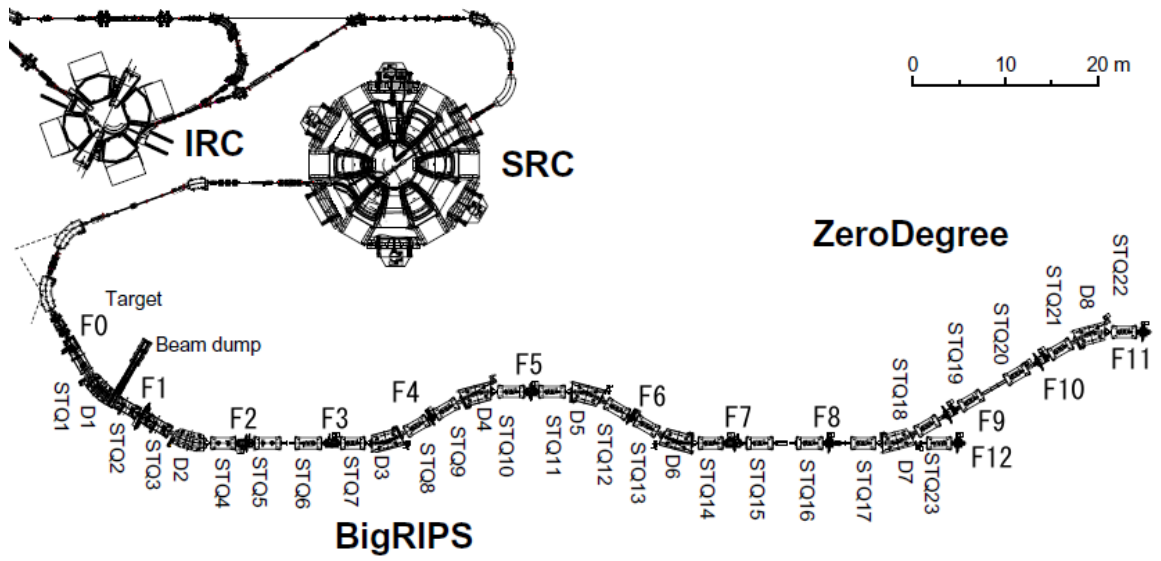


Fig. 1. Schematic layout of the BigRIPS separator shown along with the IRC and SRC cyclotrons and the ZeroDegree spectrometer. The labels STQn, Dn and Fn indicate the positions of superconducting quadrupole triplets, room-temperature dipoles and focal planes, respectively. The BigRIPS separator spans the region from the production target position (F0) to the F7 focus.

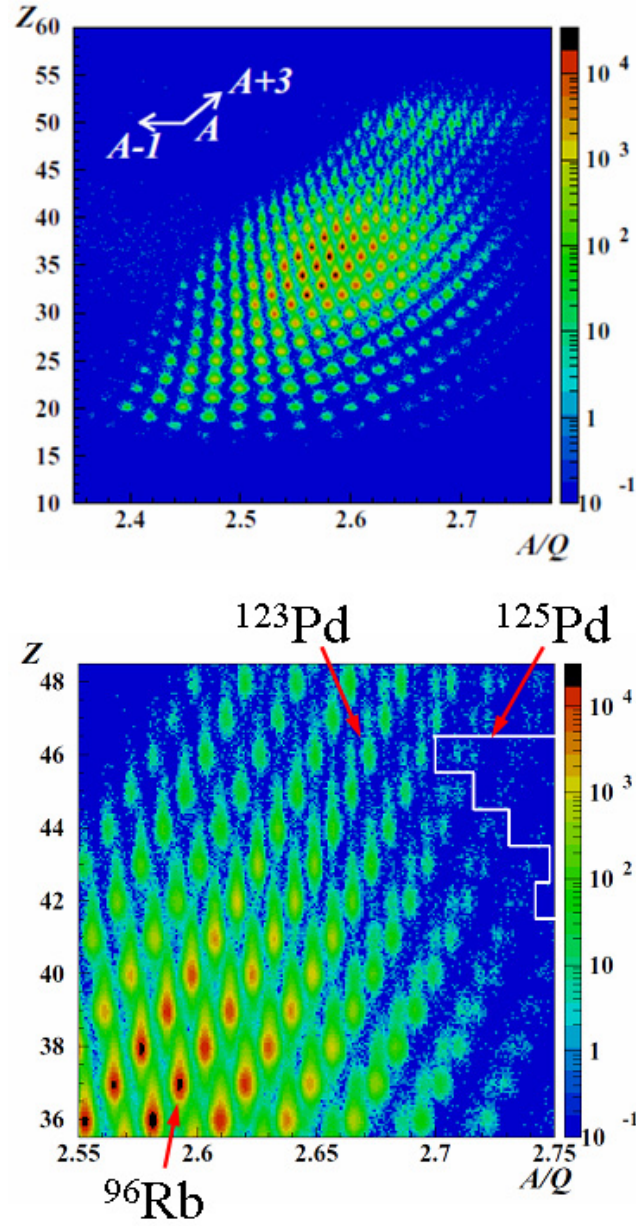


Fig. 2. (Upper) Z versus A/Q plot for fission fragments produced in the $^{238}\text{U}+\text{Be}$ reaction at 345 MeV/nucleon. (Lower) An enlarged view for the isotopes with $Z=36$ to 48, where the known limits are shown by a white line. The data was obtained with a $B\rho$ setting of 7.395 Tm and a momentum acceptance of $\pm 1\%$. The low- Z isotopes are not seen in the upper panel due to the threshold of the trigger counter.

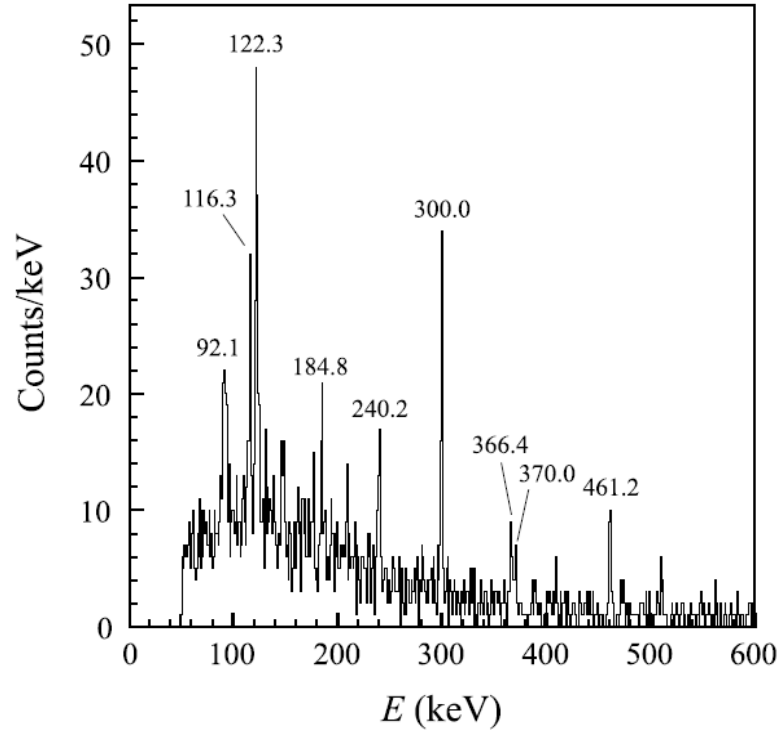


Fig. 3. Energy spectrum of delayed γ rays gated on ^{96}Rb events in the PID plot of Fig. 2. The labeled γ rays were identified as those originating from the decay of the isomeric state in ^{96}Rb (see text). Those at 122.3 keV and 300.0 keV are doublet peaks according to ref. 12.

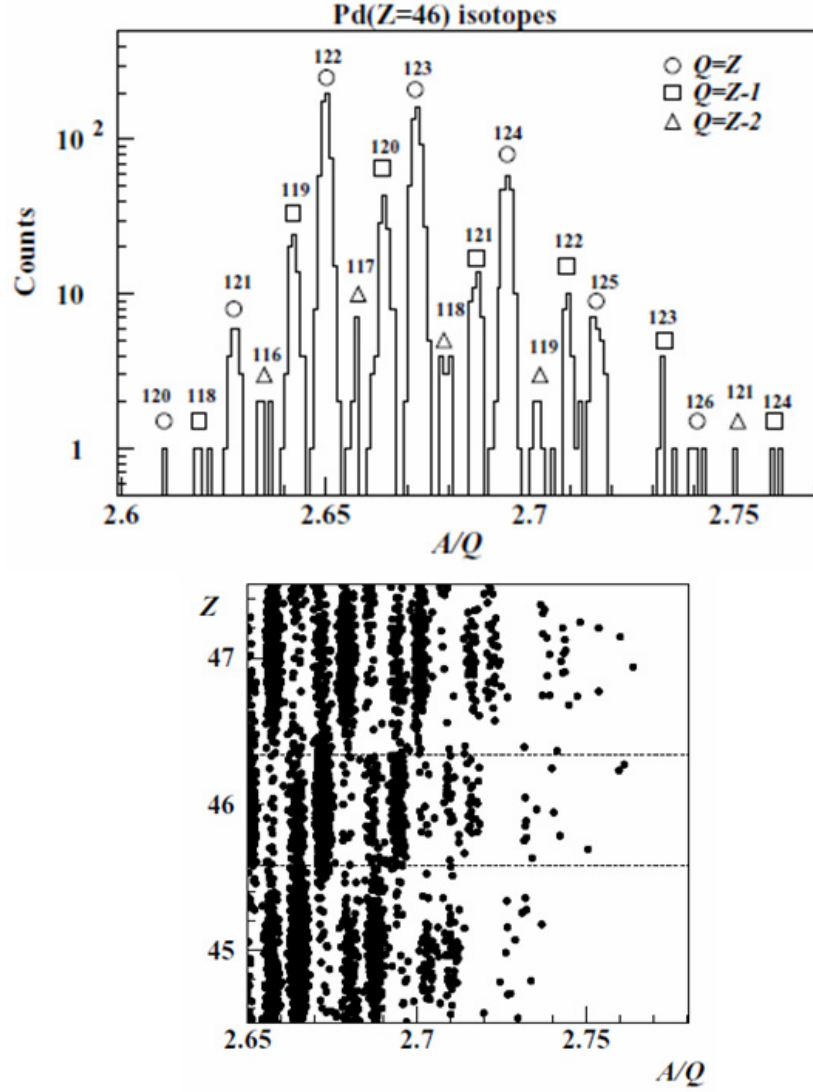


Fig. 4. (Upper) A/Q spectrum of Pd isotopes and (Lower) Z versus A/Q plot. The peaks in the upper panel are labeled by mass number and charge state. The charge states $Q=Z$, $Q=Z-1$ and $Q=Z-2$ are indicated by circles, squares and triangles, respectively. The lower panel shows the PID plot enlarged around the region of the new Pd isotopes. The horizontal dotted lines show the Z gate used to select the Pd isotopes (see text). The experimental conditions are the same as those given in Fig. 2.

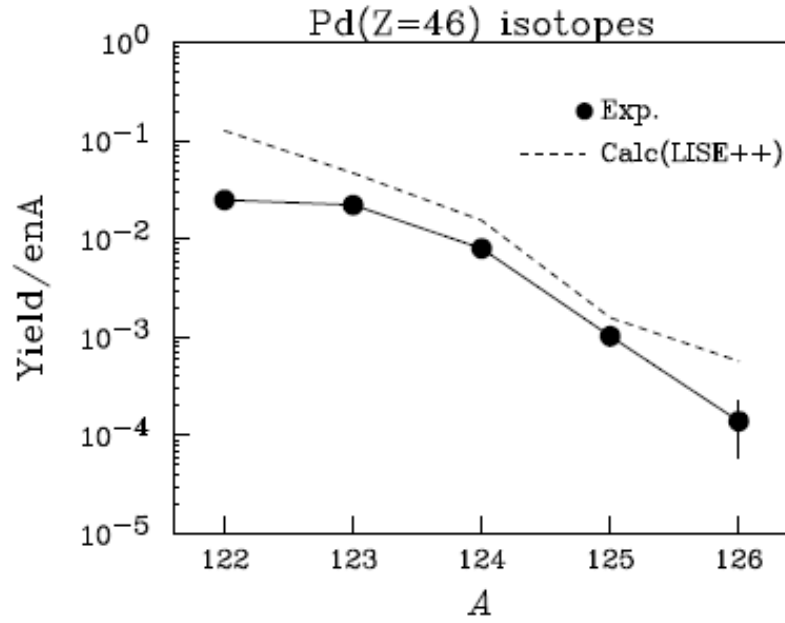


Fig. 5. Measured production rates of the Pd^{46+} isotopes shown along with the predictions from the LISE++ simulation (dashed line). The experimental conditions are the same as those given in Fig. 2.